Challenges and Opportunities for Biophotonic Devices in the Liquid State and the Solid State

A. J. Steckl, J. A. Hagen, Z. Yu, R. A. Jones, W. Li, D. Han, D. Y. Kim, and H. Spaeth

Nanoelectronics Laboratory

University of Cincinnati

Cincinnati OH 45221-0030

a.steckl@uc.edu

J. G. Grote and F. K. Hopkins US Air Force Research Laboratory Wright Patterson Air Force Base Dayton, OH

Abstract — In this paper we discuss the unique challenges and opportunities present in using biomaterials in photonics applications. We describe biophotonic materials and devices fabricated and operated in the solid state (fluorescent nanometer thin films, light emitting diodes) and in the liquid state (electrofluidic fluorescent biosensor, microfluidic switches).

Keywords - biophotonics, LED, DNA, fluorescence, nanostructures, electrofluidics.

I. Introduction

Photonics is defined as the study or application of electromagnetic energy whose basic unit is the photon, incorporating optics, laser technology, electrical engineering, materials science, and information storage and processing. Biophotonics encompasses the complex world of (a) using light to study and/or control biochemical processes; (b) using naturally occurring biopolymers (or artificially constructed facsimiles) for producing novel or otherwise improved photonic devices.

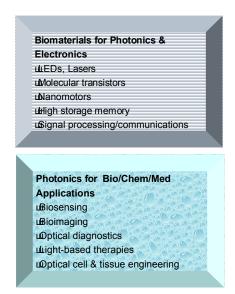


Fig. 1 The biophotonics "dry" and "wet" worlds.

In the first case, biochemistry is the driving force and biophotonic devices normally operate in the liquid state (usually aqueous solution as is the case in nature). In the second case where photonics is the goal, biophotonic devices normally operate in the solid state (consisting of multi-layer thin film structures). Examples of biomaterials for photonics (and electronics) and of photonics techniques for biochemical and biomedical applications are shown in Fig. 1. In this paper we discuss biophotonic devices that fall into each category: luminescent nanometer-thin DNA films, light emitting diodes that incorporate DNA layers, electrofluidic light wave coupled fluorescent DNA sensors, electrofluidic switches.

II. BIOPHOTONIC DEVICES

The next frontier in organic electronic and photonic devices is the use of biomaterials, either naturally occurring or artificially produced based on biological methods. Biomaterials with unusual properties not easily replicated in conventional organic or inorganic materials can provide another degree of freedom in terms of device design and produce enhancements in device performance. Furthermore, natural biomaterials are a renewable resource and are inherently biodegradable.

A. Bio-Organic Photonic Material and Light Emitting Diodes

We are investigating the use of deoxyribonucleic acid (DNA) as a photonic material and as an integral element of organic light emitting diodes (OLED). Polymer materials are of great interest for photonic applications due to their low cost and flexibility, but they generally also have higher optical loss than most inorganic semiconductor materials. Recent research suggested DNA is a good candidate in polymer materials for photonics applications. It is believed that the unique double-helix nanostructure of DNA plays an important role in intercalating dye molecules and in enhanced optical properties.

We have shown¹ that BioLEDs incorporating a DNA electron-blocking layer have significantly higher luminance and luminance efficiency compared to conventional OLEDs. For example, the luminance efficiency for a green emitting

maintaining the data needed, and c including suggestions for reducing	lection of information is estimated to ompleting and reviewing the collect this burden, to Washington Headqu ald be aware that notwithstanding an OMB control number.	ion of information. Send comments arters Services, Directorate for Info	regarding this burden estimate or rmation Operations and Reports	or any other aspect of the 1215 Jefferson Davis	nis collection of information, Highway, Suite 1204, Arlington
1. REPORT DATE 2006		2. REPORT TYPE		3. DATES COVE	tred 6 to 00-00-2006
4. TITLE AND SUBTITLE				5a. CONTRACT NUMBER	
Challenges and Opportunities for Biophotonic Devices in the Liquid State and the Solid State				5b. GRANT NUMBER	
				5c. PROGRAM ELEMENT NUMBER	
6. AUTHOR(S)				5d. PROJECT NUMBER	
				5e. TASK NUMBER	
				5f. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) University of Cincinnati, Nanoelectronics Laboratory, Cincinnati, OH, 45221-0030				8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)				10. SPONSOR/MONITOR'S ACRONYM(S)	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S)	
12. DISTRIBUTION/AVAIL Approved for publ	ABILITY STATEMENT ic release; distributi	ion unlimited			
13. SUPPLEMENTARY NO The original docum	TES nent contains color i	images.			
14. ABSTRACT					
15. SUBJECT TERMS					
16. SECURITY CLASSIFIC	17. LIMITATION OF	18. NUMBER	19a. NAME OF		
a. REPORT unclassified	b. ABSTRACT unclassified	c. THIS PAGE unclassified	ABSTRACT	OF PAGES 3	RESPONSIBLE PERSON

Report Documentation Page

Form Approved OMB No. 0704-0188 DNA BioLED reaches as high as 8 cd/A compared to 2 cd/A for a conventional OLED structure without the DNA layer.

We are exploring the use of various organic and inorganic molecules as luminescent agents in DNA. For example, the organometallic compound sulforhodamine (SRh) is part of the popular rhodamine dye family. SRh emits in the red part of the visible spectrum ranging from 600 nm to 640 nm. We are using salmon dsDNA treated² with a surfactant to change its solubility and to improve thin film quality. PL measurement was performed at 300 K with a He-Cd laser emitting at 325 nm. Fig. 2(LEFT) shows the PL emission spectrum of a SRhdoped DNA thin film. The emission peak position at 605 nm is consistent with literature report for this specific dye. To determine the doping concentration for maximum PL emission, DNA thin films with different dye concentrations were investigated. We compared the emission from the DNA:SRh films to films using a widely utilized organic host (PMMA) doped with equivalent SRh concentrations. The integrated PL emission (around the main peak at 605 nm) was used to compare the effect of SRh concentration in the two host materials. Fig. 2(RIGHT) clearly shows that the same amount of dye in DNA emits much more efficiently than in PMMA. The maximum integrated PL intensity from the DNA:SRh films is ~20× higher than the corresponding value for PMMA:SRh films.

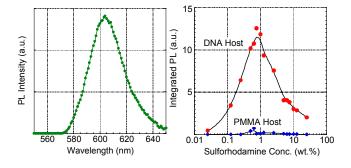


Fig. 2 Optical properties of sulforhodamine (SRh) doped DNA thin films. LEFT: Photoluminescence spectrum of DNA:SRh tun films excited by He-Cd laser at 325 nm. RIGHT: Integrated PL emission in SRh-doped DNA and PMMA films as a function of SRh concentration.

We are also investigating organic light emitting diodes that incorporate DNA thin films, so-called BioLEDs as either light emitting layers or as electron 'blocking' layers (EBL). The effect of blocking electron flow is to enhance the probability of radiative electron-hole recombination, leading to increased device luminous efficiency and luminance. Adamovich et al. first noted the use of EBL layers in OLEDs to prevent electron-hole recombination from occurring in adjacent hole transport layers (HTL)^{3,4}. DNA-based BioLEDs previously reported^{5,6} have incorporated DNA-based thin films as hosts for lumophores indicating the feasibility of the concept, but without significant improvement in device performance over conventional OLEDs. Hirata et al. have investigated⁷ the properties of DNA as a charge transport layer

in several device configurations elucidating their properties through their effect on the device current-voltage (I-V) characteristics. Organic light emitting diodes are very bright, efficient, have a relatively low fabrication cost as compared to inorganic LEDs, and have begun to gain momentum in both display and lighting applications. However, the emission spectrum is rather broad and dependent on processing conditions. The emission spectrum of rare earth ions, on the other hand, is highly monochromatic and generally independent of the host material. It is therefore interesting to investigate the properties of OLEDs and BioLEDs that combine the two types of materials and emission mechanisms.

n SO ₃	
PEDOT/PSS	W NPB ₩
, N	\bigcirc \bigcirc
0-AI-N	
	>=N-N-(
Algo	BCP
	PEDOT/PSS Alg ₃

Fig. 3 DNA-containing BioLED. LEFT: Cross-section of BioLED structure indicating the sequence and thickness of layers. RIGHT: Chemical structure of the organic components of the BioLED structure.

The device structure shown in Fig. 3 contains a 20 nm DNA thin film as electron blocking layer and a 20 nm NPB layer which serves as the host of the rare earth metal Eu, which emits a deep red color. The electroluminescence (EL) spectrum of the device is shown in Fig. 4 (LEFT). The sharp emission line at ~612 nm is due to inner shell 4f transitions in the Eu atoms, while the broad emission with dual peaks at ~450nm and 475nm, is due to the NPB layer itself. Fig. 4(RIGHT) shows the luminance of the NPB:Eu device and a baseline device (without the NPB layer and emitting from the Alq3 layer) as a function of current density. The luminance of the NPB:Eu device is clearly superior, with a maximum of 590 cd/m² at 375 mA/cm², whereas the Alq3 OLED peaks at only 45 cd/m² at 30 mA/cm².

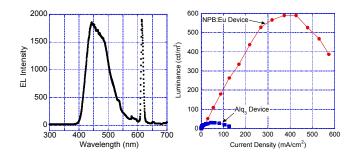


Fig. 4 Operation of DNA BioLED doped with Eu. LEFT: Electroluminescence spectrum. RIGHT: Luminance versus current density for Eu-doped BioLED and for baseline Alq₃ device.

B. Electrofluidic Fluorescent DNA Sensor

Current approaches for DNA detection have utilized either "static" methods using fluorescence techniques or "dynamic" microfluidic approaches. We are investigating a novel method for DNA detection and manipulation using light wave coupling of a pump beam into a fluid region in conjunction with electrofluidic induced transport of fluorescent DNA. As in the BioLED devices discussed in Section A, we have utilized double-stranded salmon DNA reacted with luminescent dyes. Using electrical fields in the 1-10V/cm range across aqueous DNA+dye solutions we have been able to transport the DNA to the vicinity of the waveguide, resulting in a strong fluorescence signal.

We will report the electric field dependence of the fluorescence signal intensity, rise time, decay time, as well as the effect of varying the DNA and the dye concentrations. Preliminary measurement indicates that the DNA mobility under electric field in the fluid is two orders of magnitude higher than the mobility in gel electrophoresis.

We are also investigating electrically switchable devices that can operate in the liquid state ("liquid logic") and thus can be amenable to integration with electrowetting and microfluidic devices. We have fabricated a field effect liquid state transistor. This device operates with metallic source and drain regions, a current channel formed in an electrolyte and a metallic gate.

We have demonstrated that current through this liquid state transistor can be controlled by the gate voltage. We we will report on the effect of transistor structure, materials and dimensions, as well as electrolyte concentrations on transistor gain and switching speed.

REFERENCES

- ¹ J. A. Hagen, W. Li, A. J. Steckl and J. Grote, *Appl. Phys. Lett.* **88**, 171109 (2006).
- ² E. M. Heckman, J. A. Hagen, Perry P. Yaney, J. G. Grote and F. K. Hopkins, *Appl. Phys. Lett*, **87**, 211115 (2005).
- ³ V. Adamovich, J. Brooks, A. Tamayo, A. Alexander, P. Djurovich, B. D'Andrade, C. Adachi, S. Forrest, M. Thompson, *New J. Chem*, **26**, 1171 (2002).
- ⁴ V. Adamovich, S. Cordero, P. Djurovich, A. Tamayo, M. Thompson, B. D'Andrade, S. Forrest, *Organic Electronics*, 4, 77 (2003).
- ⁵ N. Kobayashi, S. Umemura, K. Kusabuka, T. Nakahira, and H. Takashi, *J. Mater. Chem.* **11**, 1766 (2001).
- ⁶ T. Koyama, Y. Kawabe, and N. Ogata, *Proc. SPIE* **4464**, 248 (2002).
- ⁷ K. Hirata, T. Oyamada, T. Imai, H. Sasabe, C. Adachi and T. Kimura, *Appl. Phys. Lett.* **85**, 1627 (2004).

.